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# A picosecond 14.7 nm x-ray laser for probing matter undergoing rapid changes

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**Abstract.** With laser-driven tabletop x-ray lasers now operating in the efficient saturation regime, the source characteristics of high photon flux, high monochromaticity, picosecond pulse duration, and coherence are well-matched to many applications involving the probing of matter undergoing rapid changes. We give an overview of recent experiments at the Lawrence Livermore National Laboratory (LLNL) Compact Multipulse Terawatt (COMET) laser using the picosecond 14.7 nm x-ray laser as a compact, ultrafast probe for surface analysis and for interferometry of laser-produced plasmas. The plasma density measurements for known laser conditions allow us to reliably and precisely benchmark hydrodynamics codes. In the former case, the x-ray laser ejects photo-electrons, from the valence band or shallow core-levels of the material, and are measured in a time-of-flight analyzer. Therefore, the electronic structure can be studied directly to determine the physical properties of materials undergoing rapid phase changes.

## INTRODUCTION

A major goal in the pursuit of x-ray laser research has been to achieve higher efficiency, reduced size, low cost and a high repetition rate for the pumping scheme. In particular, operation in the efficient saturation regime for a tabletop-sized source is essential for applications and the future development of the field. In the last few years, the most noted examples of saturated, tabletop schemes have been based on collisional excitation pumping although the methods have been varied. The fast capillary discharge scheme produced intense Ne-like Ar lasing at 46.9 nm on the  $3p\ ^1S_0 \rightarrow 3s\ ^1P_1$  transition at high repetition rate, high average power operation [1]. The picosecond laser-driven transient schemes have been demonstrated to give in excess of 10  $\mu$ J output/pulse on the Ni-like Pd  $4d\ ^1S_0 \rightarrow 4p\ ^1P_1$  line at 14.7 nm [2]. More recently, an x-ray laser operating at 10 Hz for the Pd-like Xe ion  $5d\ ^1S_0 \rightarrow 5p\ ^1P_1$  transition at 41.8 nm was driven into saturation using 35 fs irradiation of a xenon gas cell [3]. The experimental demonstration of the second category of laser scheme, the picosecond-driven transient collisional schemes using chirped pulse amplification (CPA) laser systems, was shown first for Ne-like Ti  $3p \rightarrow 3s$  transition at 32.6 nm using 10 J of laser energy [4].

At LLNL over the last 5 years, much theoretical and experimental effort on the COMET facility has developed this picosecond, tabletop x-ray laser mainly because of the high efficiency, relatively high repetition rate and unique combination of source characteristics. Various applications, including picosecond x-ray laser interferometry of dense plasmas and probing of the electronic structure of materials, are discussed.

## APPLICATIONS OF A PICOSECOND X-RAY LASER

Typical COMET x-ray laser characteristics are listed, with the beam brightness information specifically for the Ni-like Pd line at 14.7 nm, in Table 1.

**TABLE 1.** COMET x-ray laser source parameters

Source Parameters	COMET X-ray Laser	Importance <sup>b</sup>
Laser Pump Energy (J)	5 - 10	
X-ray Laser Energy ( $\mu$ J)	25	
Photons/Shot	$2 \times 10^{12}$	<b>x</b>
Shot Rate (Hz)	0.004	
Wavelength (nm)	12 – 47	
$\Delta\lambda/\lambda$	$< 10^{-4}$	<b>x</b>
Source Size ( $\mu\text{m}^2$ )	$25 \times 100$	
Divergence ( $\text{mrad}^2$ )	$2.5 \times 10$	
XRL Pulse Duration (ps)	2 – 25	<b>x</b>
Peak Brightness, B <sup>a</sup>	$1.6 \times 10^{25}$	<b>x</b>
Average Brightness, B <sup>a</sup>	$1.3 \times 10^{11}$	

<sup>a</sup> [Units of  $\text{ph. mm}^{-2} \text{ mrad}^{-2} \text{ s}^{-1} (0.1\% \text{ BW})^{-1}$ ].

<sup>b</sup> “**x**” denotes considered important x-ray laser source parameter.

High photon flux/shot, high monochromaticity, and short pulse duration when combined with small source area and beam divergence properties of the 14.7 nm line [5] give ultra-high peak brightness  $\sim 10^{25} \text{ ph. mm}^{-2} \text{ mrad}^{-2} \text{ s}^{-1} (0.1\% \text{ BW})^{-1}$ . This assumes a 2 ps pulse duration. The range of x-ray laser pulse durations is expected to be dictated by the gain lifetime [6] and by the pulse length of the picosecond driving laser beam. A recent experiment reported the x-ray laser pulse to be 2 ps (FWHM) for a 1.3 ps-driven Ni-like Ag 13.9 nm x-ray laser [7]. Overall, the 14.7 nm peak brightness is 5 – 6 orders of magnitude higher than 3<sup>rd</sup> generation synchrotron undulator sources. The 46.9 nm fast capillary discharge x-ray laser operates at similar high peak brightness and has the further advantage, with 10 Hz repetition rate, of high average brightness,  $5 \times 10^{14} \text{ ph. mm}^{-2} \text{ mrad}^{-2} \text{ s}^{-1} (0.1\% \text{ BW})^{-1}$ . From Table 1, this is more than  $10^3$  times higher than the Pd line brightness. Third generation synchrotron

**TABLE 2.** Summary of x-ray laser applications with desired x-ray laser characteristics <sup>a</sup>

Application	Average Power	Brightness	$\Delta\lambda/\lambda$	Short Pulse <sup>b</sup>	Coherence	Tunability	Polarization	Short Wavelength
<b>Biology:</b>								
Microscopy	-	Yes	-	Yes	-	-	-	2.4 – 4.4 nm
Holography	-	Yes	-	Yes	Yes	-	-	Yes
Diffraction	-	Yes	-	Yes	-	-	-	0.1 – 1 nm
<b>Plasmas:</b>								
Interferometry <sup>c</sup>		Yes	Yes	Yes	Yes	-	Maybe	-
Non-linear Optics	-	Yes		Yes	Yes	-	Yes	-
Plasma Generation	Maybe	Yes	-	Yes/ -	Yes	-	-	-
Warm Dense Matter	Yes	Yes	Yes	Yes	-	Yes	-	0.1 nm
<b>Atomic and Molecular Physics:</b>								
Photoionization	Sometimes	Yes	Yes	Yes	-	Desirable	Sometimes	Yes/ -
Multiphoton Processes	-	Yes	-	Yes	-	Yes	-	Yes
<b>Material Science and Chemistry:</b>								
Semi-conductors	Yes	Yes	Yes	Yes	-	Yes	-	1 – 30 nm
Photoemission Spectroscopy <sup>c</sup>	-	Yes	Yes	Yes	Maybe	Yes/ No	-	1 – 60 nm
<b>Industrial:</b>								
EUVL Printing	Yes	-	-	-	-	-	-	13.5 nm
EUV Metrology	Yes	-	-	-	Yes	-	-	13 – 14 nm

Note: The symbol “-“ in row for a given application refers to characteristic that is not required or applicable.

<sup>a</sup> After Table in ref. [8].

<sup>b</sup> Short pulse defined as femtosecond to picosecond regime.

<sup>c</sup> Work described in this paper.

undulator sources still have higher average brightness of  $0.5 - 6 \times 10^{18}$  ph. mm<sup>-2</sup> mrad<sup>-2</sup> s<sup>-1</sup> (0.1% BW)<sup>-1</sup> at 50 – 10 nm, respectively. The collisional x-ray lasers have discrete wavelengths while synchrotrons are tuneable.

Table 2 summarizes some of the possible applications using x-ray lasers with the corresponding x-ray characteristics. This is based on a table published 10 years ago [8] and includes some updates and minor changes reflecting recent developments. The table is not meant to be exhaustive covering all possible applications, but lists relevant areas of interest in biology, high temperature plasmas, atomic and molecular physics, material science, chemistry and industrial semiconductors. For example, x-ray laser microscopy of biological samples first demonstrated by Da Silva et al. [9] requires the characteristics of peak brightness, short pulse duration and wavelength in the “water window”. The Extreme Ultraviolet Lithography (EUVL) project for printing semiconductor masks specifies high average power, 30 – 100 W at 13.5 nm, with high uniform spatial and constant temporal source requirements [10]. Recently, x-ray lasers have been focused at high intensity to form laser-produced plasmas [11]

Laser-driven x-ray lasers using COMET type facilities are unique sources on account of their high photon flux/pulse, high monochromaticity, and ps pulse duration in the 12 – 47 nm regime. Presently, these x-ray lasers have excellent peak brightness capability compared to other sources but the average power or brightness parameter, while still high, requires further development. Therefore, applications using high peak brightness are a good match. In addition, the short pulse duration is essential for probing transient events in high temperature plasmas, material phase changes or chemical processes. For example as shown in Table 2, x-ray laser interferometry of high temperature laser-produced plasmas, when the additional requirements of transverse and longitudinal coherence are satisfied, is an excellent application for this source. The ultra-high peak brightness is necessary to overcome x-ray self-emission from the plasma being probed. Likewise, the same x-ray source properties, high photon flux/pulse, high monochromaticity and ps pulse, are important for a surface analysis probe to study the valence band and shallow core electronic structure.

These applications are described in the next sections with both sets of experiments performed on the COMET facility. Optimization of the laser conditions for generating the saturated, 14.7 nm, Ni-like Pd x-ray laser have been detailed previously [2, 5]. For these experiments a 600 ps long pulse followed by a 6 ps short pulse beam, separated by 700 ps peak-to-peak was chosen to give optimum x-ray laser output [12].

## **PICOSECOND X-RAY LASER INTERFEROMETRY**

The first soft x-ray laser interferometry experiments were performed on the NOVA laser by Da Silva and co-workers with the 15.5 nm Ne-like Y laser using an amplitude-division Mach-Zehnder interferometer equipped with multi-layer coated thin foils as beamsplitters [13]. Large 1-mm-scale plasmas were probed to a maximum electron

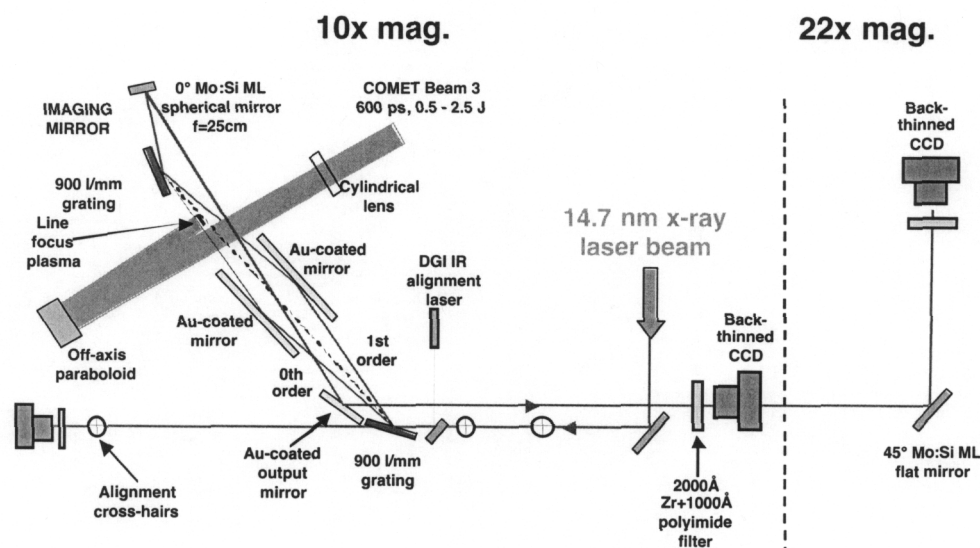


Fig. 1 Layout for interferometry experiments. Setup for 10× magnification imaging and for higher 22× imaging with additional 45° multilayer optic (to right of vertical dashed line).

density of  $2 \times 10^{21} \text{ cm}^{-3}$  and to within  $25 \mu\text{m}$  of the initial target surface. More recently, Rocca and co-workers developed a Mach-Zehnder type instrument at  $46.9 \text{ nm}$  using diffraction gratings as beam splitters [14]. Diffraction gratings are robust, use well-established technology and can be utilized for various wavelengths, as short as  $2 \text{ nm}$ . They also have a high throughput which makes them a good match to the present generation of tabletop picosecond laser-driven x-ray lasers. A version of the diffraction grating interferometer (DGI) was developed and optimized by Colorado State University for the ps Ni-like Pd  $14.7 \text{ nm}$  x-ray laser. Figure 1 shows the setup for the laser-produced plasma interferometry experiment with more details found in ref. [5]. Experiments to date have shown that even with multiple modes present in the x-ray laser beam, excellent interference fringe visibility of  $0.72 \pm 0.12$  can be achieved. Al plasmas with lengths  $0.1 - 0.5 \text{ cm}$  were irradiated by a line focus at  $10^{11} - 10^{12} \text{ W cm}^{-2}$  with a  $600 \text{ ps}$ ,  $1054 \text{ nm}$  laser pulse were probed. Two dimensional (2-D) plasma density profiles were measured and compared to 1-D, 1.5-D and 2-D hydrocode simulations [15]. Two campaigns to probe plasmas have been conducted. The first used 10× magnification imaging giving pixel limited spatial resolution of  $2.5 \mu\text{m}$  at the plasma. Lateral plasma expansion and features within  $10 \mu\text{m}$  of the target surface were observed demonstrating the effectiveness of picosecond soft x-ray probes for large millimeter-scale plasmas.

Figure 2 is an example of an interferogram for a  $0.5 \text{ cm}$  long plasma heated by a  $600 \text{ ps}$ ,  $1054 \text{ nm}$  laser pulse. A peak density of  $3 \times 10^{20} \text{ cm}^{-3}$  was measured and was limited by the detector to resolve fringe structure close to the target where the scalelengths are short. In principle, the x-ray laser interferometry technique should be able to probe

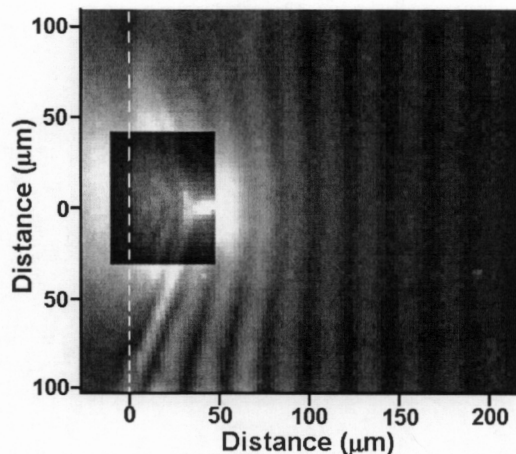


Fig. 2 A picosecond 14.7 nm x-ray laser interferogram of a 0.5 cm Al plasma recorded at 0.7ns after the peak of 0.4 J, 600 ps pulse. Laser line focus is a  $0.6 \text{ cm} \times 40\text{-}\mu\text{m}$  and target surface is at  $0 \mu\text{m}$ . Two different look up tables are used for clarity.

several orders of magnitude higher density with substantially less refraction compared to other UV or optical interferometric techniques. To address this issue, a second more recent experiment with higher 22 $\times$  magnification gave substantially improved spatial resolution of  $0.6 \mu\text{m}$ . This data is under analysis and will be reported in the near future.

## PICOSECOND X-RAY LASER PROBING OF MATERIALS

Various compact, pulsed extreme ultraviolet (EUV) and soft x-ray sources, for example higher order harmonic (HOH) [16, 17] sources for valence band and laser produced plasma (LPP) line and continuum emission [18] for core levels, have been used for the direct measurement of the electronic structure of material surfaces. Static chemical shifts in the Si  $2p$  core level electronic structure for  $\text{SiO}_2$  and  $\text{Si}_3\text{N}_4$  samples were demonstrated using 2.5 ns LPP, 255 eV x-ray pulses after integrating for  $\sim 100$  shots [18]. The HOH sources have the advantage of shorter duration since the process is driven by ultrafast pulses, e.g. less than 60 fs [16], at a high repetition rate which



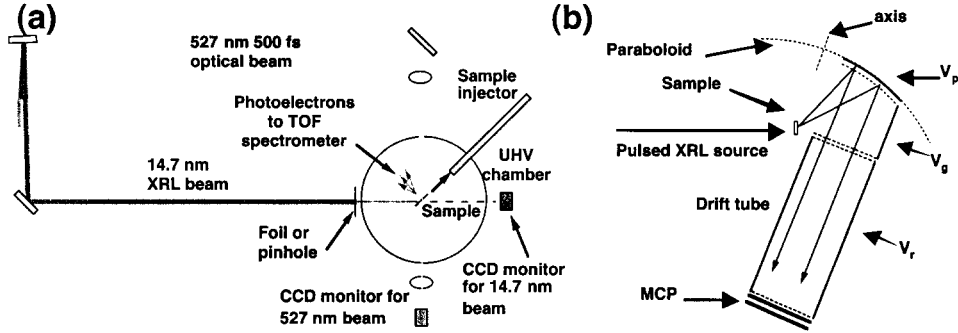


Fig. 3 (a) Layout for optical pump – x-ray laser probe of material surfaces. (b) Schematic of paraboloid mirror analyzer time-of-flight photoelectron spectrometer.

minimizes other plasma x-rays reaching the sample. A pinhole or thin filter isolates the beamline from the experimental ultra-high vacuum (UHV) chamber. The UHV chamber, operated at  $5 \times 10^{-7}$  mbar pressure, contains the sample to be probed and the photoelectron spectrometer, shown in Fig. 3(b). Photoelectrons, with a total binding energy and sample work function less than 84.5 eV photon energy ejected from the sample with a certain kinetic energy (KE), are collected by the paraboloid mirror and collimated before entering the drift tube [19]. A bias applied to the paraboloid mesh  $V_p$  and the retardation grid  $V_r$  can be used in tandem as a bandpass filter to control the energy and time of arrival of the electrons on the microchannel plate (MCP) detector. The signal is digitized using a fast 3 GHz oscilloscope. Figure 4 (a) and (b) show the time-of-flight spectra recorded for an *in-situ* sputter-cleaned Ta reference sample illuminated with  $10^9 - 10^{10}$  x-ray laser photons. The valence band and two shallow core levels ( $4f_{7/2}$  and  $4f_{5/2}$  with binding energy of 22 and 24 eV, respectively) are expected to arrive between 100 and 150 ns. Peaks corresponding to these features are observed in the spectrum sitting on a background slope of secondary photo-emission (earlier than 100 ns). The secondary emission is believed to be produced by scattered photons and higher KE electrons and will be reduced further through the use of filters

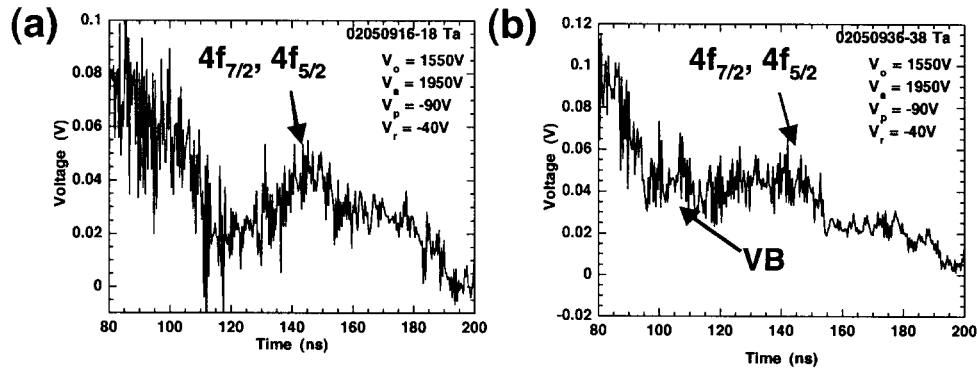


Fig. 4 (a), (b) Time-of flight photoelectron spectra from a sputter-cleaned Ta sample for two sets of x-ray laser shots. Valence band and  $4f_{7/2}$  and  $4f_{5/2}$  shallow core levels are labeled.

and baffles in the setup. In addition, electrical noise, apparent on the spectra of Fig. 4 (a) and (b), is registered on the detector and is associated with the plasma generating the x-ray laser. This presently limits the signal to noise ratio and requires averaging several shots, even though there are sufficient photoelectrons for a single shot record. This is under investigation and new results will be reported soon.

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